Role of Nanostructures in the Radiative Recombination Process in InGaN-based Light-Emitting-Diodes

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Owing to a novel two-flow metalorganic chemical vapor deposition (MOCVD) method, Nichia Chemical Industries have successfully commercialized yellow/green/blue/ultraviolet light-emitting diodes (LEDs) based on InGaN/GaN quantum-well structures [1]. The LEDs demonstrate high brightness, high quantum efficiency and long lifetime at room temperature in spite of a high density of dislocations. Although a considerable amount of researches have been done, there is still much left unknown about the radiative mechanism of the InGaN-based LEDs.

Recently many authors have recognized that the complex crystalline nanostructures and/or mesoscopic structures found in the epilayers of InGaN-based LEDs may contribute to the radiative recombination. Some authors [2, 3] presented evidences that the localized states are originated from the composition fluctuation of $In_xGa_{1-x}N$ alloys. Many experimental data are believed to support the localized or weak-localized exciton (carrier) state recombination process as the mechanism leading to spontaneous emission. On the other hand, some other authors [4, 5] suggested a picture of a highly phase segregated alloy. They stressed that the so-called InGaN alloy is really a GaN crystal containing a large number of nearly pure InN dots, and these quantum dots provide highly efficiency center for radiation recombination of excitons and carriers.

In order to give a reasonable explanation of the unique properties of the InGaN-based LEDs, we have analyzed and compared a large number of published results relating to the physical properties of the InGaN epitaxial layers. We feel that the InGaN epitaxial layer produced by Nichia Chemical Industries may be not simply a mixed crystal or an alloy with statistical composition fluctuation, it may be a sophisticated layer containing a large number of interconnected mesoscopic columnar structures that are arranged vertically on the substrate. The dislocations appear only in the edge region of the columns. Figure 1 illustrates schematically the structure of an idealized InGaN-based LED under our consideration. The average diameter of the mesoscopic crystalline columns is estimated about 100∼1000 nm corresponding to the dislocation density of $10^8 \sim 10^{10}/cm^2$.

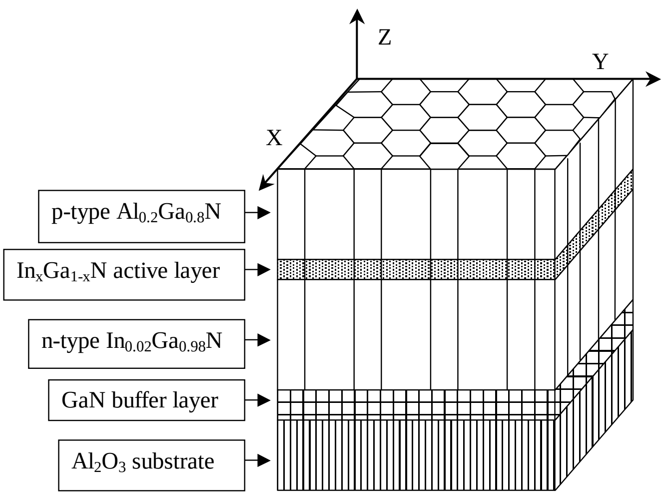

Figure 1: Mesoscopic structures of an idealized InGaN-based LED.

Based on the x-ray diffraction measurements published by many study groups, we can suppose reasonably the lattice structures in the self-formed mesoscopic columns are good, but the concentration distributions of the elements are inhomogeneous. There may be InN nanostructures in the mesoscopic columns. We guess that, in the epitaxial growth process, free InN molecules tend to deposit on the top of InN molecules that are just in the top layer for the favor of the enthalpy. If the growth condition is not changed, InN chains will grow along the growth direction, usually the c axis of the crystal. The diameters of the InN chains are dependent on the average indium concentration and the growth condition. In an $In_xGa_{1-x}N$ ($x < 0.05$) barrier layer, the diameter of the InN chain is estimated only a few atoms in breadth because of the very low indium concentration, but the length of the chain is mostly the same of thickness of the barrier layer. So that, it is better to described the InN chain structure in the barrier layer as a quantum wire system. The situation in the active layer is different. The size of the In-rich region has been estimated in the order of ten to hundred nm on the plane, but the thickness of the active layer is only a few monolayers. So that the In-rich nanostructure in the active layer is better described as a quantum disk system. Figure 2 shows schematically the quantum confinement structures in an idealized mesoscopic column.

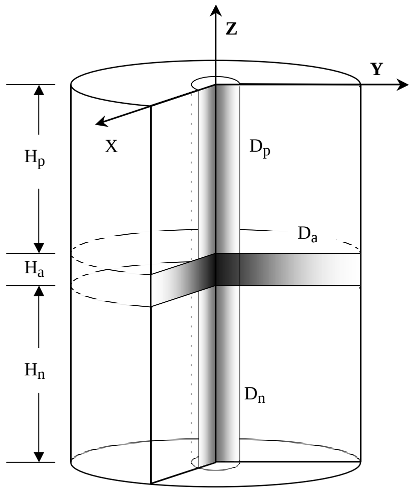

Figure 2: Schematic nanostructures in an idealized InGaN crystalline column. H_n, H_a and H_p are the thickness of the n-type barrier, the active layer and the p-type barrier, respectively. D_n and D_p are the diameter of the quantum wire in the barriers, D_a is the diameter of the quantum disk in the active layer.

Based on the ideal model mentioned above, we study the properties of InGaN-based LEDs and epitaxial films. It is revealed that some important properties, such as the high quantum-efficiency and the Stokes shift of the LEDs, may originate from quantum confinement effects of self-formed InN quantum wires in the GaN layers and InN-rich quantum disks with central concentration distribution in the InGaN active layer. The present theoretical work gives a reasonable and systematical explanation of the radiative recombination mechanism of InGaN-based LEDs and epitaxial layers.

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